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Carbonaceous Aerosols in the Industrial Era

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Carbonaceous aerosols are increasingly recognized as an important atmospheric constituent. These small atmospheric particles are predominately soot produced by incomplete combustion of fossil fuels, biofuels, and outdoor biomass that generally form through condensation of vaporized organic matter [*Chylek et al.*, 2003]. However, biogenic emissions from trees, other vegetation, and animals are also sources of carbonaceous aerosols. Elemental carbon, in the form of graphite, is the main cause of the blackness of soot; it absorbs sunlight strongly and almost uniformly across the solar spectrum. However, the graphite seldom is pure carbon, instead involving varying proportions of other atoms. Furthermore, the carbonaceous aerosols include an enormous variety of organic compounds of carbon.

Carbonaceous aerosols are usually divided, rather arbitrarily, into categories of black carbon (BC) and organic carbon (OC). This division is based on the aerosol optical properties, not on their chemistry. The carbonaceous aerosols that absorb visible radiation strongly are defined as BC, and the remaining carbonaceous aerosols as OC. OC aerosols may absorb visible light weakly, with this absorption usually increasing toward ultraviolet wavelengths.

Aerosols Affect Global and Regional Climate

It has been suggested that the radiative forcing by BC contributes substantially to global warming [*Hansen et al.*, 2000; *Jacobson*, 2001], although the warming effect may be reduced by accompanying OC and the indirect effects of these aerosols on cloud brightness and cloud cover [*Penner et al.*, 2003]. Carbonaceous aerosols also affect regional climate patterns [*Ramanathan et al.*, 2001], for example, possibly contributing to a trend toward increased rainfall in south China and drought in north China [*Menon et al.*, 2002]. These aerosols also decrease the albedo of snow and ice surfaces, and in this way might contribute to global warming and the worldwide melt-back of glaciers, sea ice, and permafrost [*Hansen and Nazarenko*, 2004].

The history of atmospheric carbonaceous aerosols is needed to investigate their possible role in past climate change. Quantitative knowledge of the contributions of fossil fuels, biofuels, and outdoor biomass to carbonaceous aerosols would be needed for any strategies to mitigate aerosol effects, including aerosol effects on human health, agricultural productivity, and atmospheric visibility, as well as on climate.

Carbonaceous aerosol history is difficult to estimate because soot emissions depend sensitively on combustion technology, unlike, for example, CO_2. Because aerosol lifetimes are typically only several days, their global distribution is inhomogeneous; thus, data are required from many locations. The aerosol size and vertical distributions are needed because they influence the aerosol climate effects.

Aerosol Models and Emission Scenarios

Global aerosol chemical transport models are a central tool in strategies for defining carbonaceous aerosols over the industrial era because of the aerosol spatial inhomogeneity and the sparseness of measurements. The validity of aerosol emission scenarios and the capability of chemical transport models must be repeatedly tested and improved using a variety of data. Satellite data and ground-based networks can test recent emissions and modeling processes. Emission histories can be checked via ice core records and other innovative approaches.

Emission scenarios depend on uncertain fuel use histories and even more uncertain "emission factors." Information is needed on transitions from one fuel use sector to another; for example, from household-based to power plant burning of coal. Even within a sector, emission factors change with time; as, for example, with the transition from household stoves to less polluting central heating. Reduction of aerosol emissions as diesel technology improved is known for specific cases, but it is difficult to define this on a global basis.

Laboratory measurements of emission factors are needed. Accurate global measurements of aerosol amount and composition today, compared with aerosol models fed by specified sources, will provide a check on current emissions. Historical emission inventories can be checked via several data sources discussed

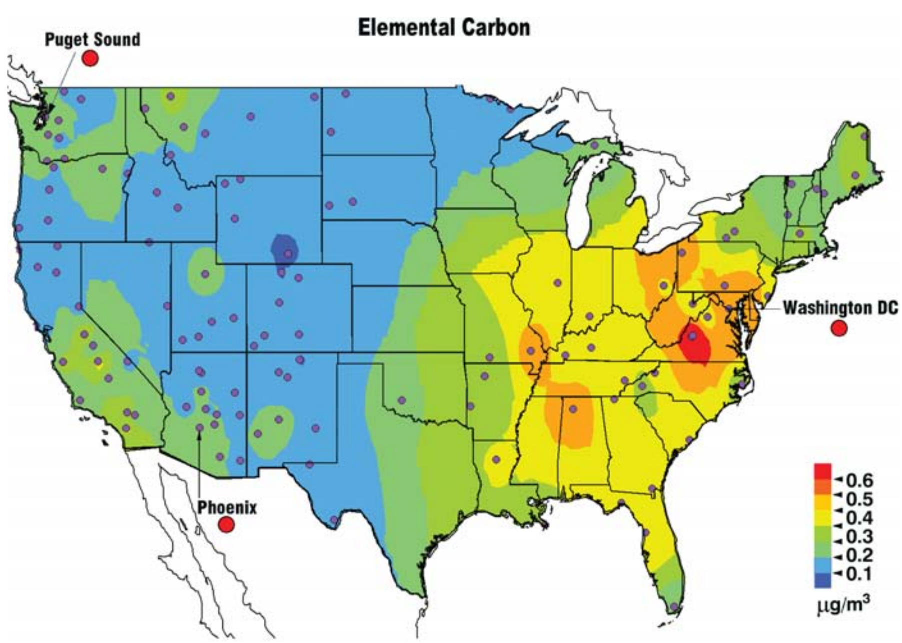

Fig. 1. Average BC concentrations (ug/m³) for 2001 were provided by the IMPROVE network [Malm et al., *2004]. Isopleths are for concentrations in rural areas. Urban amounts are greater.*

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below. In these tests, the possible variation of the OC/BC ratio over time should be addressed, because the climate effect of carbonaceous aerosols depends strongly on that ratio. Also, it is important to know how biomass burning has changed over time, an issue that might be investigated via ice, lake, and bog core records from a variety of places.

Aerosol Surface Deposition

Typical BC amounts in ice cores vary from 0.1–0.5 parts per billion by weight (ppbw) at the South Pole, to 1–5 ppbw in Greenland, to 5–50 ppbw at middle latitude sites in the northern hemisphere. In the Alps, BC increased from about 20 ppbw in the early 1800s to 70 ppbw in the second half of the 20th century. BC histories could also be obtained in the Himalayas, East Africa, western Canada, and South America from ice cores. Lake and bog cores provide widespread sites, although local fires affect these cores more than in the case of remotely located ice cores. However, high-resolution analyses of many cores in one region have the potential to offer a useful BC history at the decadal, centennial, and millennial scales [*Clark and Royall*, 1996]. Because lake and bog cores are scattered throughout the terrestrial biosphere, they could be used to provide BC histories in key locations.

Studies of aerosol histories could be advanced by support for: a working group to develop standard BC and OC measurement procedures, including a reference sample [*Schmidt et al*., 2003]; new approaches such as radio carbon (radioactive isotope C-14) analysis to determine fossil and biomass proportions; worldwide measurements of BC content in snow surfaces; ice core and sediment core analysis at many global sites; and local and regional atmospheric measurements in conjunction with deposition measurements, for example, at Jungfraujoch.

Air sampling networks provide valuable aerosol data for recent decades. The most extensive network in the United States, the Interagency Monitoring of Protected Visual Environments (IMPROVE), has 54 sites in national parks [*Malm et al*., 2004], with data from 1989 to the present. IMPROVE includes chemical analyses for BC, OC, sulfate, and nitrate. Figure 1 shows the distribution of BC in 2001, ranging from 100–200 $\mu g/m^3$ in the mountain states to 500 $\mu g/m^3$ in the eastern United States. The seasonal and inter-annual variations of BC suggest that fossil fuels are the largest source of BC in the United States, but biomass burning contributes substantially. The IMPROVE record, although as yet only 14 years long, provides valuable measures of the OC/BC ratio and trends of BC and OC [*Malm et al*., 2004].

Filter measurements of "black smoke" have been made in Europe and the United States for several decades. These data can be converted, with possible error up to a factor of 3, to BC absolute amount. Despite this uncertainty, the black smoke records have the potential to yield the relative change of BC over time; in some cases, over several decades.

More quantitative information could be obtained with modern chemical analyses of archived filter collections. Archived filters can be subjected to optical analyses for BC, and in some cases, OC analyses may still be possible. Custodians of available samples need to be identified. Interpretation of old filters would be aided by parallel sampling now with original sampling substrates, as well as with present-day sampling and analysis protocols.

Atmospheric Transparency and Satellite Data

Atmospheric transmission and visibility records provide indications of changes in aerosol amount and aerosol absorption, although broadband data can be confused by changes in ozone, water vapor, and even clouds in the case of all-sky radiation. Mauna Loa broadband transmission data, for example, suggest a small, long-term (42-yr) increase of aerosols over the Pacific Ocean [*Dutton and Bodhaine*, 2001]. Comparison of clear sky direct and diffuse solar radiation at the surface in Germany and the United States indicates that over the period 1960 to 1990, there was an increase of absorbing aerosols relative to scattering aerosols [*Liepert and Tegen*, 2002].

Additional information might be obtained from a search for solar radiation measurements from old field campaigns; for example, measurements by C. Junge in the Himalayas in the 1950s and by *Mani et al*. [1969] in India in the 1960s. (Mani et al. described the aerosol layer as "a great brown sea lapping against the Himalayas," crediting the phrase to Reid Bryson.) Additional information might also be obtained from analysis of historic Brewer measurements (ozone measurements made at two wavelengths in the ultraviolet in clear sky), and through collection of historic transmission data for many sites, analogous to the Global Energy Balance Archive data [*Ohmura et al*., 1998], but for clear sky.

Satellite measurements provide near-global data that will increasingly constrain global aerosol transport models and emission inventories as the capabilities of the satellite data improve and the satellite records become longer. The longest satellite records, covering 2 decades, have low accuracy for aerosol amount, yet the spatial and inter-annual variations usefully constrain aerosol models. Current satellites provide a distinction between coarse and accumulation mode aerosol sizes, as well as lidar measurements of the vertical profile of aerosols. Future satellite measurements with wide-angle, full-spectral polarimetry will provide more information on aerosol single-scatter albedo and size distribution, thus allowing aerosol speciation and improved differentiation of carbonaceous aerosols based on refractive index.

Other Approaches

Other suggested approaches include analysis of historical photographs, which cover the period since the invention of photography in about 1850. Photographs throughout the world are available and could provide measures of visual range. Photographs near sources such as smokestack plumes might help constrain historical emission estimates.

Another possibility is analysis of ancient stone crusts (archaeometry), consisting of analysis of the black crust deposited on ancient stone monuments. Investigators in France and Italy have obtained data on black carbon in this way, specifically showing that the air in southern France (1180–1636) and northern Italy (1530–1887) was polluted by wood combustion from medieval to pre-industrial times. Black particles increased in the 19th and early 20th centuries, and fuel oil deposits were found after 1950.

Carbonaceous aerosols may be the largest source of uncertainty in analyses of climate change during the industrial era. Great improvement in understanding of carbonaceous aerosol history is possible, but it requires an innovative combination of global aerosol models; improved emission inventories; increasingly precise satellite data; enhanced capabilities at ground station networks; and exploitation of archival records, including those in ice and lake cores.

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Space Weather Research Elucidates Risks to Technological Infrastructure

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"Space weather" refers to electromagnetic and particle conditions in space near the Earth that may produce problems for space-borne and ground-based technological systems, and which may even be dangerous to humans. Explosions on the Sun are the primary origin of space weather phenomena. The solar wind carries the effect to the Earth, and the interaction with the geomagnetic field affects the magnetosphere, which has a sharp boundary on the dayside and a long tail in the nightside. The plasmaphysical coupling between the magnetosphere and the ionosphere is also important. The entire chain, starting from the Sun and ending at geomagnetic storms observed at the Earth's surface, involves complicated processes whose better understanding will lead to possibilities of avoiding harmful impacts. In space weather research, galactic cosmic rays have to be considered as a secondary source.

The activity of the Sun follows the 11-year sunspot cycle. Figure 1 shows the variation of the sunspot number, and the bars indicate the occurrence of geomagnetic storms. The diamonds on the top show times of reported ground-based troubles caused by space weather. The problems tend to occur near sunspot maxima, but technological systems can also be disturbed during sunspot minima.

Transportation and Space Flight

Space weather affects all stages of a space flight. The lift-off of spacecraft may be delayed due to difficulties in communication produced by solar X-ray bursts. Increased radiation can affect astronauts, preventing space walks. Future manned interplanetary flights should be timed for low doses of galactic cosmic rays, and shielding against particles from the Sun is necessary. Spacecraft face risks from four sources: hot plasma electrons produce surface charges on satellites; high-energy electrons penetrate the spacecraft and can disturb or destroy electronics; particles and UV radiation produce long-term degradations of satellite equipment, for example, solar cells; and enhanced UV radiation causes upward expansions of the atmosphere, which creates increased orbital drag for satellites. Abrupt decreases of the altitude of satellites have led to losses of communication with satellites.

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Aviation

Secondary cosmic rays are the main source of radiation exposure for aircraft crews, passengers, and on-board electronics. The route and the altitude have a decisive influence on the amount of radiation. Routes at high latitudes entail greater exposure due to the deeper penetration of galactic cosmic rays into the atmosphere near the geomagnetic poles. Dose variations during the 11-year solar cycle are studied in the projects Dosimetry of Aircrew Exposure to Radiation during Solar Maximum and Air Crew Radiation Exposure Monitoring, both funded by the European Union (EU). Other studies also look into whether the aircraft type has an influence on the dose [*Oksanen*, 1998]. Effects due to cosmic rays have led to radiation protection laws for aviation in Europe.

Microchips on modern aircraft are susceptible to radiation; equipment containing CMOS (Complementary Metal Oxide Semiconductor) semiconductors exhibit a 100% increase in the soft error rate (SER) when the flight altitude is changed from 9 km to 20 km [*Ziegler and Srinivasan*, 1996].

Railways

Geomagnetically induced currents (GIC) are driven by an induced geoelectric field in railway equipment during space weather storms. In railway systems, the knowledge of GIC magnitudes, the associated voltages, and their impacts is still quite poor. The only well-documented GIC problem on railways occurred in Sweden during a geomagnetic storm in July 1982; traffic lights turned red even though there was no train coming, because an induced voltage cancelled the operational voltage, which should only be short-circuited by a train. But there is reason to believe that some of the many failures classified as "unknown" on railways are due to space weather.

Cosmic rays can also affect trains by passing through and disturbing semiconductor devices. Such impacts occurred in the German high-speed train Inter City Express.

Telecommunication and Navigation

The first space weather effects on technological systems were observed by telegraph operators more than 150 years ago [*Boteler et al.*, 1998]. At times, their devices became inoperable, while at other times, they operated even without a battery. The geoelectric field drives GIC in communication cables.

A large geomagnetic storm in March 1940 broke telecommunication devices in northern

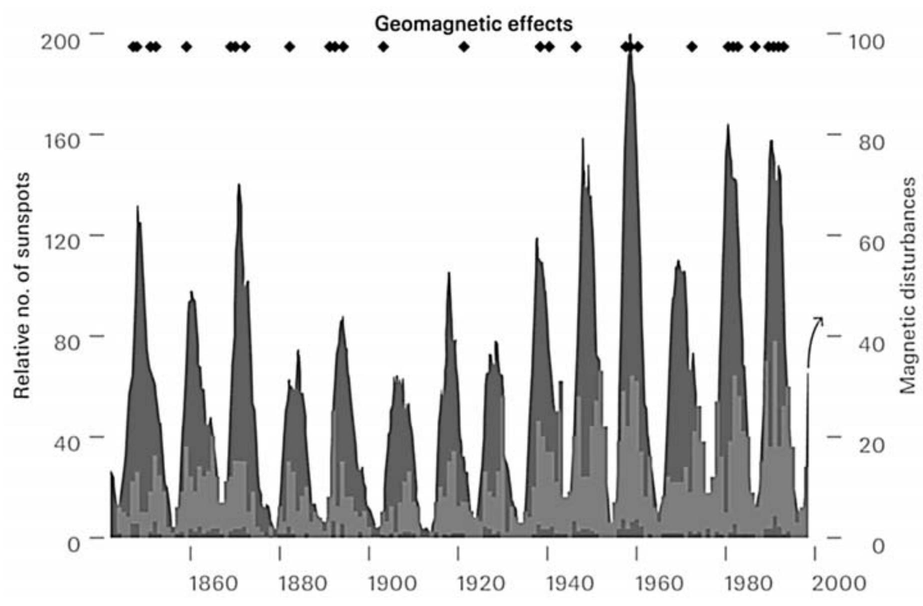

Fig. 1. The curve shows the variation of the sunspot number during the last ~150 years. The bars indicate the occurrence of geomagnetic storms; two different shades of gray refer to different intensity levels of the storms. The diamonds on the top of the figure are the times of reported problems in ground-based technological systems due to geomagnetic effects (modified by David Boteler from Boteler et al. *[1998]).*